SUB-ÅNGSTRÖM STABILIZATION OF AN X-RAY FREE ELECTRON LASER OSCILLATOR AND NUCLEAR RESONANCE METROLOGY*

B.W. Adams†, K.J. Kim‡, ANL, Argonne, IL 60439, USA

Abstract

A scheme is described to length-stabilize the cavity of an x-ray free-electron-laser oscillator (XFELO) by locking one of its longitudinal modes to the narrow nuclear resonance line of ^{57}Fe at 14.4 keV. With a cavity thus stabilized, a standing-wave pattern can be maintained over hours, to be probed by another sample of ^{57}Fe in a meter-long scan to compare the nuclear-resonant wavelength with a known optical standard. This will improve by at least four orders of magnitude the accuracy at which the ^{57}Fe resonance wavelength can be measured. The technique can be refined for other, narrower resonances such as ^{181}Ta (6.2 keV, 75 peV), opening up precision x-ray metrology for technological applications and fundamental physics, such as, e.g., experimental quantum gravity.

INTRODUCTION

Metrology is at the core of modern physics and technology, both for fundamental studies, and practical applications. A striking example for the latter is the global positioning system (GPS), which relies on the accuracy of atomic clocks and shows the relevance of general-relativistic corrections for the Earth's gravitational field. Atomic clocks can measure time - and thus also length - to an accuracy of a few times 10^{-16}. Following current trends [1], this figure will probably improve incrementally over the next few years. Progress is made through careful control of environmental perturbations and ensemble averaging for good signal statistics of the clock phase. Efforts are underway to replace the current, microwave-based clocks with optical ones. These are less susceptible to stray fields, and tick at a higher rate for faster phase accumulation. The next natural step in this progression is to use x-ray nuclear resonances for their even higher frequencies and the high degree of isolation of nuclei within their host electronic shells. For nuclear-resonant length and time standards to be useful, one has to be able to relate them to the existing optical ones. This would require means of optical coupling the two. Unlike in the optical regime, where lasers provide intense coherent light, and nonlinear optics can couple modes, x-ray optics has to date been mainly restricted to low-coherence and linear-optical cases. The reasons for this are mainly the lack of intense, coherent, and stable x-ray sources, and the weakness of nonlinear-optical effects at x-ray frequencies. The former of these can now be addressed with the recently studied x-ray free-electron oscillator (XFELO) [2, 3, 4], which can provide x-ray beams of high coherence and high intensity (measured in terms of the number of photons per mode). The present proposal has two part to it - the first showing a way to further improve the coherence characteristics of an XFELO through cavity stabilization, and the other proposing a way of relating x-ray to optical wavelengths without the use of nonlinear optics. Although the high intensity of an XFELO beam could presumably lead to nonlinear-optical effects, this is currently too much an uncharted territory to be the basis of a proposal. Instead, the identities of well-defined pieces of matter in an interferometer are exploited for their interactions with visible light and with x-rays.

* This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

† adams@aps.anl.gov

‡ kwangje@aps.anl.gov

XFELO

An XFELO combines an x-ray optical cavity made of near-backscattering Bragg crystals with an x-ray free-electron laser to produce a constant stream of intense, coherent, and wavelength-tunable pulses. The XFELO pulses are copies of the same pulse circulating in the cavity. Thus, if the pulse spacings can be stabilized to much better than the wavelength, then the XFELO output will be a frequency comb similar to that in mode-locked optical lasers. Unlike in the optical case, the x-ray comb does not cover a full octave in frequency, and thus cannot be used in self-referencing schemes. As a specific example optimized for the present proposal, an XFELO with the following parameters will be considered: The cavity is tuned to the 14.4-keV nuclear resonance of ^{57}Fe, and is formed by four diamond crystals oriented to the (337) reflection ($2\Theta = 162°$). It has a round-trip length of L=90 m, corresponding to a pulse spacing of $T = L/c = 0.3\mu$s, and a longitudinal-mode spacing of 3 MHz in the frequency comb, i.e., 14 neV on the energy scale. Electrons of 7 GeV in bunches of 25 pC in an rms length of 0.5 ps will be used for for the FEL. The intracavity peak power of this XFELO is 35 MW, and the output has $3 \cdot 10^8$ photons in a 3-meV bandwidth. This amounts to about 1000 photons per mode and pulse. The small-signal gain is 0.5, and the round-trip loss (including outcoupling) is 26%. [1].

The linewidth of the modes in the frequency comb is given by fluctuations in the cavity length, spontaneous undulator emission, and by noise in the FEL gain process. The former are caused by seismic ground motion and vibrations due to local machinery. These, and ways to mitigate them, have been studied extensively by the gravitational-wave-detector community, and their experience is used here. Spontaneous undulator emission into the FEL lasing mode has a power of about 10^{-8} of the intracavity radiation. This leads to a random phase jump of about 10^{-4}

[1] We thank R.R. Lindberg for these parameters

ISBN 978-3-95450-123-6

radians in each pulse, while fluctuations in the modulus are attenuated in the equilibrium of gain and loss in the FEL. Gain-related noise depends on fluctuations in the electron-beam parameters (energy, emittance, etc.). In a rough estimate, one may expect a phase shift of $\pi/2$ for a 100-percent gain variation. To make the gain-related noise not exceed the spontaneous-emission noise, bunch-to-bunch gain fluctuations must therefore be kept at the 10^{-4} level.

To model the spectral broadening in the frequency comb, we apply a generic model for the electric field $E(t)$ in the XFELO output, writing

$$E(t) = \sum_{n=1}^{N} e^{i\psi_n} E_0(t - nT) \tag{1}$$

where $E_0(t)$ is the electric-field amplitude of a single pulse. Its exact shape determines the spectral width of the emission, which needs not be specified here because only a sub-μeV part of the spectrum will be used. ψ_n is the phase error of the n-th pulse, for which we assume uncorrelated shot-to-shot phase fluctuations ϕ_n around a zero mean, and cumulative phase errors φ_k.

$$\psi_n = \phi_n + \sum_{k=0}^{n} \varphi_k \tag{2}$$

Both are modeled here as independent Gaussian random variables with the rms values

$$\langle \phi_k \phi_\ell \rangle = \sigma_\phi^2 \delta_{k\ell}, \quad \langle \varphi_k \varphi_\ell \rangle = \sigma^2 \delta_{k\ell}, \quad \langle \varphi_k \phi_\ell \rangle = 0 \tag{3}$$

This model leads to an output spectral power density of the form

$$\left\langle \left| \tilde{E}(\omega, \omega_T) \right|^2 \right\rangle = N F(\sigma_\phi^2, \sigma^2, \omega/\omega_T) \left| \tilde{E}_0(\omega) \right|^2, \tag{4}$$

where N is the number of pulses included in the average, and

$$F(\sigma_\phi^2, \sigma^2, x) = \left(1 - e^{-\sigma_\phi^2} + e^{-\sigma_\phi^2} \frac{\sinh(\sigma^2/4)\cosh(\sigma^2/4)}{\sinh^2(\sigma^2/4) + \sin^2(\pi x)} \right) \tag{5}$$

with $x = \omega/\omega_T$.

For small σ and σ_ϕ, and x close to an integer n, eq. (5) reduces to $(\sigma/2)^2/((\sigma/2)^2 + (\omega - n\omega_T)^2)$, i.e., a Lorentzian line. This is in qualitative agreement with the Lax model of a noise-driven harmonic oscillator [5]. In the ultimate limit, with cavity-length and gain fluctuations suppressed below the spontaneous-emission value of $\sigma = 10^{-4}$, the linewidth is 10^{-8} of the 14-neV line spacing in the comb, i.e., less than 100 atto-eV.

CAVITY STABILIZATION

Elementary, Single-Mode Stabilization

The stabilization scheme [6] is shown schematically in Fig. 1. A sample of nuclear-resonant material, ^{57}Fe with a resonant energy of $\hbar\omega_R = 14.4$ keV, and a linewidth of $\hbar\Gamma$ =5 neV in our example, is placed into the output of the XFELO. This linewidth is always subject to some inhomogeneous broadening. About 8 neV can be obtained in nonmagnetic samples of high chemical symmetry such as iron hexacyanide. This is sufficiently narrow to resolve the 14-neV mode spacing of the XFELO, so that one mode can be locked to the nuclear resonance in a feedback scheme where a piezoelectric actuator changes the cavity length. Following resonant absorption of a photon from the

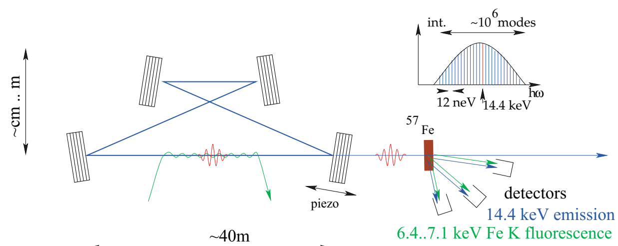

Figure 1: XFELO with crystals A..D forming an x-ray optical cavity. Radiation leaving by the output coupler (A) excites ^{57}Fe to resonant absorption, the strength of which is measured to keep one longitudinal mode on resonance.

XFELO, a ^{57}Fe nucleus will re-emit this photon, or eject a 1s electron of its host atom. The latter process is followed by emission of one x-ray fluorescent photon at an energy of 6.4 or 7.05 keV. Both the the resonantly emitted photons, and the x-ray fluorescence can be detected in a 4π solid angle as a measure of the absorption strength, i.e., the overlap of a longitudinal mode in the XFELO output with the ^{57}Fe resonance. The only exception is the exact forward direction where the radiation detector would be overloaded with the almost a million times stronger non-resonant radiation. The optical thickness of the sample must be chosen smaller than 1 to avoid the necessity of self-absorption corrections, radiation trapping, or superradiant emission into this forward direction [7], which would be useless anyway due to superradiant line broadening. Assuming an optical thickness of 0.1, about 100 photons will be absorbed from each pulse at resonance. Then, with Poisson statistics of the absorption process, 100 pulses in a 30-μs time interval are needed for a 1-percent accurate determination of the resonant signal. With these estimates, the cutoff frequency of the feedback loop is 30 kHz. The feedback loop does not stabilize the cavity length in an absolute sense like an independent optical interferometer would do. Rather, it both compensates for the effects of ground motion, etc., and changes the cavity length to compensate for the effects of phase diffusion from the gain-related noise. This way the XFELO output is kept at resonance with the absolute frequency standard of the nuclear resonance.

We now need to compare the cutoff frequency with the noise contributions. Phase diffusion follows a root-of-time law, so with 10^{-4} radians phase jumps per pulse, a 1-percent phase change will take about 10000 pulses, or 3 ms.

ISBN 978-3-95450-123-6

This is slow enough for the feedback loop to handle. Seismic noise varies considerably by location. An empirical formula to describe ground motion $x(f)$ over the frequency f that is typical for quiet sites selected for gravitational-wave detectors is [8]

$$x(f) \approx \frac{10^{-9}m}{\sqrt{Hz}} \left(\frac{10Hz}{f}\right)^{-2} \quad (6)$$

To find the amplitude of ground motion within a time interval τ, we take the square root of the integral of the spectral power density, i.e., the square of eq. (6),

$$\sqrt{\left(\frac{10^{-9}m}{\sqrt{Hz}}\right)^2 (10Hz)^4 \int_{1/\tau}^{\infty} \frac{df}{f^4}} < 8.5 \cdot 10^{-13}m \quad (7)$$

to find that $\tau = 6 \cdot 10^{-4}$ s yields a motion of 1% of the x-ray wavelength. We conclude that a 30-kHz feedback loop can easily compensate for the ground motion to better than 1% of the x-ray wavelength.

Using Multiple Modes

The stabilization scheme as described above would take photons out of the longitudinal mode that is to be used in nuclear-resonant applications. Given the low optical thickness of 0.1 that is discussed above, the loss may seem tolerable, but more serious for applications may be interference of the XFELO output with delayed emission from the stabilizer sample. A simple remedy for this problem is to move the stabilizer sample a constant velocity to make it resonant with another mode through the Doppler shift. This technique is well known, and is used in Mössbauer spectroscopy to scan energy spectra. With ^{57}Fe, the resonance shifts by 48 neV (10 linewidths) at a velocity of 1 mm/s. Therefore, the stabilizer sample has to move at 0.3 mm/s to skip one mode. This velocity need not even be particularly accurate because errors enter the absolute resonance with a factor Γ/ω_R.

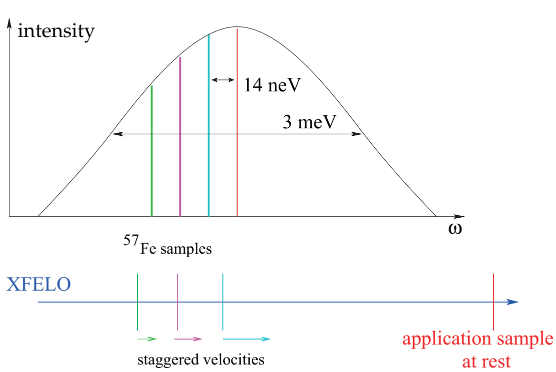

Figure 2: Multiple samples tuned to to different longitudinal modes of the XFELO to avoid interference at the application sample, and to boost signal strength.

The feedback-signal strength may be boosted by use of several stabilizer samples, each velocity-tuned to a different longitudinal mode, see Fig. 2. One may also use velocity tuning to obtain a feedback signal with information on the sign of the error. In this scheme, shown schematically in Fig. 3 for three samples, one of them is tuned to where its mode should lie, while the two others are at slightly lower and slightly higher velocities than that for full tuning to their modes. The difference of the latter two then provides the sign of the error.

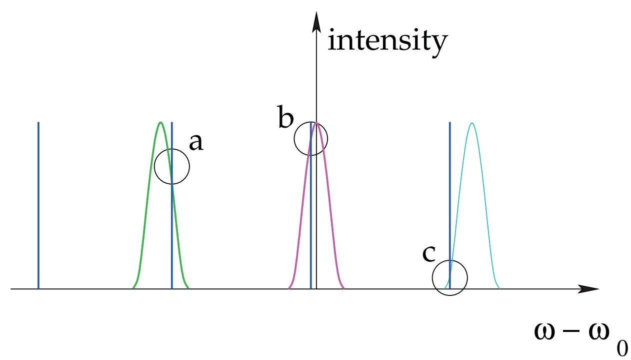

Figure 3: Using velocity tuning to obtain an error signal with sign. One of three samples (magenta resonance curve) is velocity-tuned to the peak of a longitudinal mode, another (green resonance curve is tuned at a slightly lower frequency than "its" mode, and the third (cyan curve) is tuned at a slightly higher frequency. The signal "b" should be at it peak value (here slightly off), and the difference a-c shows that the cavity length needs to be reduced to tune to slightly higher frequencies.

RELATING AN X-RAY WAVELENGTH TO OPTICAL STANDARDS

In order to enable precision metrology based on nuclear resonances, we need to relate the x-ray resonant wavelength to optical length standards. To that end, the output of the XFELO is back-reflected into itself to generate a standing-wave pattern. Unlike the XFELO cavity, where the requirement of minimal loss restricts the choice of Bragg reflector material, the standing-wave generation can incur some loss. A crystal material suitable for exact back-reflection can thus easily be found. Corresponding to the ^{57}Fe resonance is a coherence length of about 50 meters, so the standing-wave pattern extends over more than 20 m. This standing-wave pattern is then probed by nuclear-resonant absorption in a scan over a distance of several meters. To get a sufficient signal strength, one may use many ^{57}Fe atoms arrayed periodically with a lattice parameter matching the standing-wave period, or one may focus, as shown in Fig. 4, and use a single nucleus. The former method poses some challenges because the lattice-parameter match has to be sufficiently good to amount to much less than an Ångström over the thickness of the lattice, and because the lattice planes have to be aligned to be highly parallel to the standing waves. We will there-

ISBN 978-3-95450-123-6

fore discuss the option of a single nucleus in a focus. As shown in Fig. 4, a pair of Fresnel zone plates focuses the beam from the XFELO to a 50-nm waist, and re-collimates it. The back-reflected beam undergoes the same focusing. A single nucleus of ^{57}Fe is placed into this focus, embedded in a stiff matrix of a material of light elements, such as diamond. The entire assembly of zone plates and probe nucleus travels along the standing-wave pattern under optical-interferometric control. For every displacement of $\lambda_R/4 = 0.21$Å the resonant fluorescent signal from this nucleus changes from strong to weak, or vice versa. With

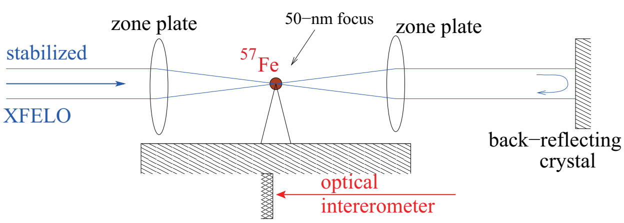

Figure 4: Scanning an x-ray standing-wave pattern with an assembly of two confocal x-ray zone plates focusing to 50 nm, and re-collimating, and a nucleus of ^{57}Fe placed into this focus.

a resonant absorption cross section of $2 \cdot 10^{-18}$ cm^2 [9], about 1000 photons per pulse and mode, and $3 \cdot 10^6$ pulses per second, a single nucleus in the 50-nm focus will absorb about 100 photons per second. This is sufficient for probing the standing waves provided the wavelength is maintained stable for the duration of the scan. It is not necessary to probe every single one of the roughly 10^{11} interference fringes per meter because pre-existing knowledge of the wavelength can be applied for sparse sampling.

Alternatively to the frequency-domain picture of standing waves at the resonant frequency (superposed by nonresonant waves), one may also apply a time-domain approach where subsequent pulses in both incident, and returning directions hit the probe nucleus, and interfere with nuclear-dipole oscillations due to previous pulse excitations, reinforcing, or diminishing them depending on the phasing of the pulse wavepackets relative to the previous pulses.

In a model similar to the one presented for the XFELO output, the response of resonant nuclei to this phased-pulse excitation can be calculated to find the contrast of the nuclear response to the intensity modulation of the standing waves. The result is shown in Fig. 5 for different values of ΓT, i.e., the pulse repetition rate in units of the inverse nuclear-resonant linewidth Γ.

OUTLOOK

The scheme described here can be applied to other nuclear-resonant species. Some of these, such as ^{181}Ta, or ^{45}Sc, have much sharper resonances than ^{57}Fe. Especially the latter could far surpass the accuracy of atomic clocks as time and length standards. Applications of a nuclear-resonant XFELO could be found in diverse fields, such as

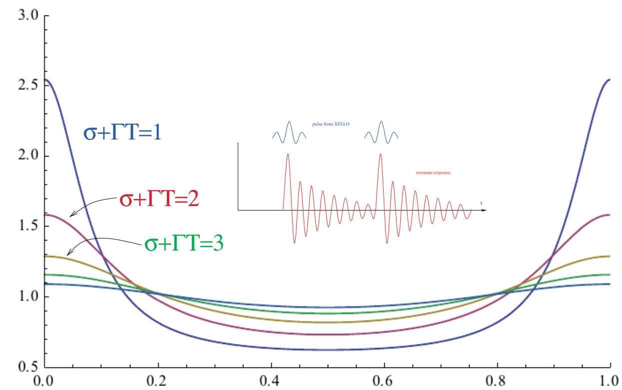

Figure 5: Contrast of nuclear-resonant absorption for different values of ΓT. The insert shows repeated in-phase excitation of nuclear-resonant oscillations.

local measurements of the gravitational potential through the gravitational red-shift. This would help generate a density map of bedrock in applications such as natural-resource exploration. Other applications may lie in using the large momentum and high spatial resolution of x-rays in fundamental quantum physics for studies of the collapse of the quantum wavefunction of mesoscopic objects.

ACKNOWLEDGMENT

We thank Stanley Whitcomb for discussions on cavity-length stabilization in gravitational-wave detectors, and Ryan Lindberg for working out the parameters of the XFELO in our example.

REFERENCES

[1] NIST. `http://www.nist.gov/pml/div688/grp50/primary-frequency-standards.cfm`, 2012.

[2] R. Colella and A. Luccio. Proposal for a free electron laser in the x-ray region. *Opt. Comm.*, 50:41–44, 1984.

[3] Kwang-Je Kim, Yuri Shvydko, and Sven Reiche. A proposal for an x-ray free-electron laser oscillator with an energy-recovery linac. *Phys. Rev. Lett.*, 100:244802, 2008.

[4] Kwang-Je Kim and Yuri V. Shvyd'ko. Tunable optical cavity for an x-ray free-electron-laser oscillator. *Phys. Rev. Sp. Top., Acc. and Beams*, 12:030703, 2009.

[5] Melvin Lax. Classical noise. V. noise in self-sustained oscillators. *Phys. Rev.*, 160:290–307, 1967.

[6] B.W. Adams and K.-J. Kim. Sub-Ångström stabilization of an x-ray free-electron laser oscillator for nuclear resonance metrology and fundamental physics. *manuscript submitted*, 2012.

[7] M.O. Scully. Correlated spontaneous emission on the Volga. *Laser Physics*, 17:635–646, 2007.

[8] Saulson. `http://cgwa.phys.utb.edu/Files/Events/29_630_Saulson_0614_Vibration_Isolation.pdf`.

[9] H. Fraunfelder. *The Mössbauer Effect*. W.A. Benjamin, New York, 1963.

ISBN 978-3-95450-123-6